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(54) Title of Invention:

FUNGICIDE COMPOSITION FOR AGRICULTURAL AND HORTICULTURAL USES

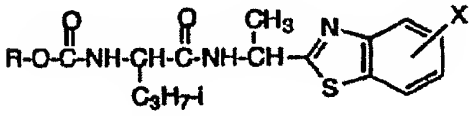
(57) Abstract:

[Problem] To provide a fungicide composition for agricultural and horticultural uses, causing no adverse effect on crops but having a broad range of fungicidal spectra at a low dosage.

[Solution] A fungicide composition for agricultural and horticultural uses, containing an amino acid amide derivative and a fungicidal compound for agricultural and horticultural uses as active components.

[Claims]

[Claim 1] A fungicide composition for agricultural and horticultural uses, comprising, as active components, a kind of amino acid amide derivatives represented by the following formula:



[1]

[wherein R denotes a lower alkyl group, and X denotes a hydrogen atom, a halogen atom, a methyl group or a methyl group] and a fungicidal compound selected from the group of compounds consisting of zinc ion-coordinated manganese ethylenebisdithiocarbamate, manganese ethylenebisdithiocarbamate, zinc ethylenebisdithiocarbamate, zinc propylenebisdithiocarbamate, N-(trichloromethylthio)cyclohexa-4-en-1,2-dicaroboxyimide, tetrachlorophthalonitrile, 3-chloro-N-(3-chloro-2,6-dinitro-4-α, α, α-trifluoromethyl-2-pyridine)amine, 2,3-dichloro-4-fluorophenylmaleimide, 1-(2-cyano-2-methoxyiminoacetyl)-3-ethylurea, aluminum tris(ethylphosphonate), cupper bis(quinoline-8-olate), cupper hydroxide, methyl-N-(2-methoxyacetyl)-N-(2,6-xylyl)-DL-alaninate, 2-methoxy-N-(2-oxo-1,3-oxazolidine-3-il)acetamide-2',6'-xylidine, methyl-N-phenylacetyl-N-2,6-xylyl-DL-alaninate, methyl-N-(2-furyl)-N-(2,6-xylyl)-DL-alaninate, (E,Z)-4-[3-(4-chlorophenyl)-3-(3,4-dimethoxyphenyl) acryloyl]morpholine, N-(trichlromethylthio)phthalimide, propyl-3-(dimethoxyamino)propylcarbamate hydrochloride, (RS)-2-(2,4-dichlorophenyl)-1-(1H-1,2,4-triazol-2-il)hexane-2-ol, (cis, trans)-3-chloro-4-[4-methyl-2-(1H-1,2,4-triazol-1-ilmethyl)-1,3-dioxolane-2-il]phe nyl-4-chlorophenylether, (E)-4-chloro-α, α, α-trifluoro-N-(1-imidazol-1-il-2-propoxyethylidene)-o-toluidine, 2,4'-chloro-α-(pyrimidine-5-il)-

benzohydrylalcohol, and 1,1'-iminodi(octamethylene)diguanidium tris(dodecylbenzenesulfonate) (iminoctazine dodecylbenzensulfonate).

[Claim 2] A fungicide composition for agricultural and horticultural uses according to Claim 1, wherein the fungicidal compound for agricultural and horticultural uses is a different compound.

[Claim 3] A fungicide composition for agricultural and horticultural uses according to Claim 1, wherein R in the amino acid amide derivatives represented by the formula [1] denotes an alkyl group and X denotes a halogen atom.

[Claim 4] A fungicide composition for agricultural and horticultural uses according to any of Claims 1~3, wherein R in the amino acid amide derivatives represented by the formula [1] is a propyl group or a butyl group.

[Claim 5] A fungicide composition for agricultural and horticultural uses according to any of Claims 1, 3 and 4, wherein an isomer in the amino acid of the amino acid amide derivatives represented by the formula [1] is of the L-form.

[Claim 6] A fungicide composition for agricultural and horticultural uses according to any of Claims 1 and 3~5, wherein an isomer in the amine section of the amino acid amide derivatives represented by the formula [1] is of the R- or RS-form.

[Claim 7] A fungicide composition for agricultural and horticultural uses according to any of Claims 1 and 3~5, wherein an amino acid derivative represented by the formula [1] and one or more kinds of compounds selected from the group of fungicidal compounds for agricultural and horticultural uses are blended in a weight ratio of 1:05 ~ 1:1000.

[Detailed Description of Invention]

[0001]

[Field of Invention] The present invention relates to a fungicide composition for agricultural and horticultural uses, which contains an amino acid amide derivative and a fungicidal compound for agricultural and horticultural uses as active components.

[0002]

[Prior Art] Amino acid amide derivatives are disclosed in Laid-Open German Patent 4321897, but compounds to be used according to the present invention, wherein the amine section in the amino acid amide derivatives is of the R-form, are novel substances not having been specifically disclosed in said German Patent. Fungicidal compounds for agricultural and horticultural uses are either commercially available or known compounds as fungicides for agricultural and horticultural uses, and these compounds are known as disclosed in Pesticides Handbook (1994) published by Japan Plant Protection Association, Kumiai Pesticide Manual (1995) published by National Federation of Agricultural Cooperative Association, SHIBUYA INDEX (1993) published by said National Federation, etc.

[0003]

[Problem to be Solved by Invention] Various kinds of fungicides for agricultural and horticultural uses are commercially available and have been used to prevent and control disease injuries to useful plants, but some of these agents have poor post-infection fungicidal activities against pathogenic fungi (poor therapeutic activities). Also, due to the repeated application of a fungicide, chemical-resistant fungi have become flourishing in late years so as to reduce the plant protection activity of some chemical agents, and some chemical agents have now become substantially

useless. Moreover, when a plant is infected with a certain disease, various disease injuries can be also caused in combination, but some of commercial agents have narrow fungicidal spectra. Phycomycetes causing downy mildew of cucumbers, late blight of tomatoes, downy mildew of grapes, late blight of potatoes, etc. are one of pathogenic causes particularly difficult to control. Even an effective chemical agent for controlling some other disease injuries can be poorly effective in the Phycomycetes control, and there are chemical agents which cannot be used for controlling both the Phycomycetes and other pathogenic diseases.

[0004] The purpose of the present invention is to provide a fungicide composition for agricultural and horticultural uses having a broad fungicidal spectrum for controlling the disease injuries of useful plants at a low dosage, particularly, a fungicide composition for agricultural and horticultural uses capable of exhibiting excellent post-infection fungicidal activity with a fungicidal compound for agricultural and horticultural uses which has low post-infection fungicidal activity (low therapeutic effect), exhibiting the outstanding activity of controlling chemical-resistant fungi with a fungicidal compound for agricultural and horticultural uses having a control activity lowered by chemical-resistant fungi, and exhibiting the extremely high activity of controlling Phycomycetes with a fungicidal compound for agricultural and horticultural uses having the poor or no activity of controlling Phycomycetes.

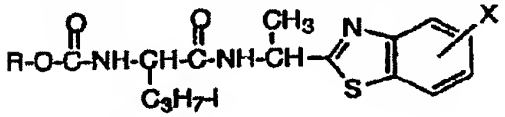
[Solution of Problem]

[0005] As a result of intensive investigations to make a more excellent chemical agent out of commercially available fungicides for agricultural and horticultural uses, the present inventors have found that a fungicide composition according to the present invention can have excellent

fungicidal activity to a broad range of pathogenic fungi at a low dosage, particularly an agent having excellent fungicidal activity after pathogenic infection at a low dosage, an agent exhibiting outstanding activity of controlling chemical-resistant fungi, and also an agent having the combined activity of controlling Phycomycetes (Zygomycota and Oomycota) can be obtained by the combined use of an amino acid amide derivative represented by the formula [1] and a fungicide composition for agricultural and horticultural uses, thus leading to the completion of the present invention.

[0006] A fungicide composition for agricultural and horticultural uses according to the present invention comprises an amino acid amide derivative represented by the formula [1] and a fungicide composition for agricultural and horticultural uses, as active components.

[0007] Namely, the present invention relates to a fungicide composition for agricultural and horticultural uses, comprising a kind of amino acid amide derivatives represented by the formula [1]:



[1]

[0008] (wherein R denotes a lower alkyl group, and X denotes a hydrogen atom, a halogen atom, a methyl group or a methyl group) and

[0009] one or more fungicidal compounds selected from the group of compounds consisting of (A) zinc ion-coordinated manganese ethylene-bisdithiocarbamate (Manzeb), (B) manganese ethylenebisdithiocarbamate (Maneb), (C) zinc ethylenebisdithiocarbamate, (Zineb), (D) zinc propylene-bisdithiocarbamate (Propineb), (E) N-(trichloromethylthio)cyclohexa-4-en-1,2-dicaroboxyimide (Captan), (F) tetrachlorophthalonitrile (TPN), (G) 3-chloro-N-(3-chloro- 2,6-dinitro-4-α, α, α-trifluoromethyl-2-pyridine)

amine (Flurazinam), (H) 2,3-dichloro- 4-fluorophenylmaleimide (Fluoroimide), (I) 1-(2-cyano-2-methoxyiminoacetyl)-3-ethylurea (Cymoxanil), (J) aluminum tris(ethylphosphonate) (Fosetyl aluminum), (K) cupper bis(quinoline-8-olate) (oxyquinoline cupper), (L) cupper hydroxide (cupper (II) hydroxide), (M) methyl-N-(2-methoxyacetyl)-N-(2,6-xylyl)-DL-alaninate (Metraxyl), (N) 2-methoxy-N-(2-oxo-1,3-oxazolidine-3-il)acetamide-2',6'-xylidine(Oxadixyl), (O) methyl-N-phenylacetyl-N-2,6-xylyl-DL-alaninate (Benalaxyl), (P) methyl-N-(2-furyl)-N-(2,6-xylyl)- DL-alaninate (Furalaxyl), (Q) (E,Z)-4-[3-(4-chlorophenyl)-3-(3,4-dimethoxyphenyl) acryloyl]morpholine (Dimethomorpf), (R) N-(trichlromethylthio)phthalimide (Folpet), (S) propyl- 3-(dimethoxyamino)propylcarbamate hydrochloride (Propamocarb hydrochloride), (T) (RS)-2-(2,4-dichlorophenyl)-1-(1H-1,2,4-triazol-2-il)-hexane-2-ol (Hexaconazol), (U) (cis, trans)-3-chloro-4-[4-methyl-2-(1H-1,2,4-triazol-1-ilmethyl)-1,3-dioxolane-2-il]phenyl-4-chlorophenyleth er, (Difenaconazol), (V) (E)-4-chloro-α, α, α-trifluoro-N-(1-imidazol-1-il-2-propoxyethylidene)-o-toluidine (Triflumizol), (W) 2,4'-chloro-α-(pyrimidine-5-il)-benzhydrylalcohol (Fenarimol), and (X) 1,1'-iminodi-(octamethylene)diguanidium tris(dodecylbenzenesulfonate) (iminoctazine dodecylbenzensulfonate), as active components..

[0010] A lower alkyl group in R of the amino acid amide derivative represented by the formula [1] denotes a linear or branched group having 1~6 carbon atoms, examples of which may include methyl, ethyl, n-propyl, isopropyl, n-butyl, isobutyl, sec-butyl, tert-butyl, pentyl, 1-methylbutyl, 2-methylbutyl, 3-methylbutyl, 2,2-dimethylpropyl, 1,1-dimethylpropyl, 1-ethylpropyl, hexyl, isohexyl, etc. A halogen atom in X denotes an atom of fluorine, chlorine, bromine, iodine, etc.

[0011] A fungicide composition for agricultural and horticultural uses according to the present invention preferably comprises an amino acid amide derivative represented by the formula [1] wherein R is a lower alkyl group and X is a halogen atom, and a compound wherein R is a propyl or butyl group, particularly R is an isopropyl or tert-butyl group is preferable. Also, an isomer in the amine section of the amino acid amide derivative is preferably of the L-form. A preferable isomer in the amine section of the amino acid amide derivative is either of the R- or RS-form, and particularly preferable is the R-form.

[0012] A fungicide composition for agricultural and horticultural uses according to the present invention, which comprising a mixture of an amino acid amide derivatives represented by the formula [1] and a fungicidal compound for agricultural and horticultural uses have very excellent fungicidal activity, and exhibits excellent activity, at a low dosage, to control various diseases caused by fungi belonging to Plasmodiophoromycetes, Chytridiomycetes, Zygomecetes, Oomycetes, Ascomycetes, Deuteromycetes and Basidiomycetes and other pathogenic fungi. The fungicide composition exhibits particularly excellent activity at a low dosage of controlling various diseases caused by Pseudoperonospora, for example, Pseudoperonospora cubensis, Phytophthora, for example, Phytophthora infestans and Phytophthora infestans, for example, Plasmopara viticola, etc.

[0013] The present invention is featured of the excellent post-infection activity. Namely, fungicides A~L and Q~S as used according to the present invention cannot exhibit satisfactory activity of controlling disease injuries when used at a low dosage. Plants are always subjected to the risk of pathogenic fungi in farms where the plants are cultivated. A timing of

chemical treatment for controlling disease injuries is often after the infection by pathogenic fungi, and fungicidal compounds A~H, J~L, R and S have a very low post-infection activity. Fungicidal compounds I and Q have to be used at high dosages to achieve a sufficient fungicidal effect and prolonged fungicidal power. However, when these fungicidal compounds A~L and Q~S are used in combination with an amino acid amide derivative represented by the formula [1], a chemical agent capable of exhibiting excellent post-infection fungicidal activity and prolonged effectiveness at a low dosage can be obtained.

[0014] A fungicide composition for agricultural and horticultural uses according to the present invention also exhibit excellent activity to chemical-resistant fungi. Namely, fungicidal compounds M~P are suffering from a lowered controlling activity due to the repeated application of these compounds leading to the flourishing of pathogenic fungi resistant to these chemicals, but when these fungicidal compounds are used in combination with an amino acid amide derivatives represented by the formula [1], excellent fungicidal activity to the chemical-resistant fungi can be exhibited.

[0015] Fungicidal compounds T~X can be hardly expected to be useful to control Phycomycetes, but when these fungicidal compounds are used in combination with an amino acid amide derivatives represented by the formula [1], agents having combined fungicidal activity also to Phycomycetes can be obtained.

Amino acid amide derivatives represented by the formula [1] to be used according to the present invention are shown in Table 1. Incidentally, compound numbers are referred in the following description.

[0016] [Table 1]

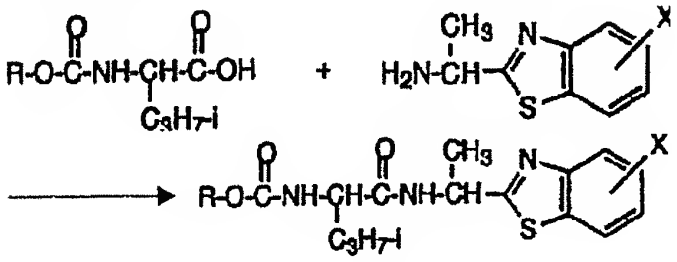
compound no.	R	Xn	isomer in amino acid	isomer in amine section	melting point or reflective index
1	t-C4H9	H	L	R	
2	t-C4H9	H	L	RS	66-67
3	i-C3H7	H	L	R	180-184
4	i-C3H7	H	L	RS	187-189
5	t-C4H9	6-Cl	L	R	133-134
6	t-C4H9	6-Cl	L	RS	128-131
7	i-C3H7	6-Cl	L	R	194-195
8	t-C4H9	6-F	L	R	128-129
9	t-C4H9	6-F	L	RS	132-134
10	i-C3H7	6-F	L	R	167-168
11	i-C3H7	6-F	L	RS	188-191
12	t-C4H9	6-CH3	L	R	122-124
13	t-C4H9	6-CH3	L	RS	
14	i-C3H7	6-CH3	L	R	190-192
15	i-C3H7	6-CH3	L	RS	
16	t-C4H9	6-OCH3	L	R	145-147
17	t-C4H9	6-OCH3	L	RS	
18	i-C3H7	6-OCH3	L	R	205-207
19	i-C3H7	6-OCH3	L	RS	
20	CH3	6-F	L	R	
21	C2H5	6-Cl	L	R	

[0017] An amino acid amide derivative represented by the formula [1] can be prepared according the below describe preparation process (A) or (B).

[0018] [Preparation Process (A)]

[0019] [Chem. Formula 2]

Formula [2] Formula [3]



Formula [1]

(wherein R and X denote the same as defined above).

[0020] An amino acid amide derivative represented by the formula [1] to be used according to the present invention can be prepared by the reaction of an amino acid derivative represented by the formula [2] or a compound wherein its carboxylic group is activated with amines represented by the formula [3] with or without the presence of a catalyst and/or a base.

[0021] In this reaction, examples of a compound having an activated carboxyl group of the amino acid derivative represented by the formula [2] may include acid halogenides such as acid chloride, etc., acid anhydrides formed by the dehydrating condensation of 2 molecules of the amino acid derivative represented by the formula [2], mixed acid anhydrides constituted by an amino acid derivative represented by the formula [2] and other acid or o-alkyl carbonate, etc., activated esters such as p-nitrophenyl esters, 2-tetrahydropyranyl esters, 2-pyridyl esters, etc., and the like.

[0022] The reaction may be run by using a condensation agent such as N,N'-dicyclohexylcarbodimide, carbonyldiimidazole, 2-chloro-1,3-dimethyl-imidazolium chloride, etc.

[0023] A solvent usable in the reaction may be any solvent as far as it does not inhibit the reaction, and its examples may include hydrocarbons such as pentene, hexane, heptane, cyclohexane, petroleum ether, ligroin, benzene, toluene, xylene, etc., halogenated hydrocarbons such as dichloromethane, dichloroethane, chloroform, carbon tetrachloride, chlorobenzene, dichlorobenzene, etc., ethers such as diethylether, diisopropylether, ethyleneglycol dimethylether, tetrahydrofuran, dioxane, etc., ketones such as acetone, methylethylketone, methylisopropylketone, methylisobutylketone, etc., acetic esters such as methyl acetate, ethyl acetate, etc., nitriles such as acetonitrile, propionitrile, benzonitrile, etc., aprotic polar solvents such as dimethylsulfolane, dimethylformamide, sulfolane, etc. and solvent mixtures combining members selected from these solvents.

[0024] A base generally used in such a type of reaction can be used. Its examples may include alkali metal hydroxides such as sodium hydroxide, potassium hydroxide, etc., alkali earth metal hydroxides such as calcium

hydroxide, etc., alkali metal carbonates such as sodium carbonate, potassium carbonate, etc., organic bases such as triethyleneamine, trimethyleneamine, dimethylaniline, pyridine, N-methylpyridine, 1,5-diazabicyclo[4,3,0]non-5-ene (DBN), 1,8-dizabicyclo-[5,4,0]-undecene-7-ene (DBU), etc. Preferable examples are tertiary amines such as triethyleneamine, pyridine, N-methylpyridine, etc.

[0025] Examples of a catalyst may include 4-dimethylaminopyridine, 1-hydroxybenzotriazol, dimethylformamide, etc. A reaction temperature is within the range of –70°C~100°C, preferably within the range of –60°C~40°C. A reaction time is preferably 1~20 hours.

[0026] A compound represented by the formula [2] as a raw material can be prepared, for example, as N-tert-butoxycarbonyl-L-valine by the reaction of L-valine and di(tert-butyl)dicarbonate in the presence of sodium hydrogen carbonate. Also, it may be prepared as N-isopropoxycarbonyl-DL-valine by the reaction of DL-valine and isopropyl chloroformate in the presence of sodium hydrogen carbonate. These are already known processes [refer to Methoden der Organischen Chemie, vol.15, no.2, p.2 (Georg Thieme Verlag Stuttgart, 1974); Chemistry of the Amino Acids, vol.2, p.891 (John Willey & Sons, N.Y., 1964); and Journal of the American Chemical Society, vol.79, p.4686 (1957)].

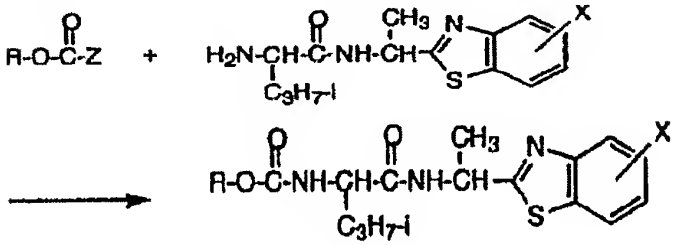
[0027] Among these compounds having an activated carboxyl group in the amino acid derivative, for example, a mixed acid anhydride can be prepared by the reaction of an amino acid derivative represented by the formula [2] and pivaloyl chloride in the presence of organic bases. Also, p-nitrophenyl ester can be prepared by the reaction of an amino acid derivative represented by the formula [2] and p-nitrophenol in the presence of a condensation agent. These are known processes [refer to Methoden der

Organischen Chemie, vol.15, no.2, p.2 (Georg Thieme Verlag Stuttgart, 1974); Chemische Berichte, vol.38, p.605 (1974); Journal of the American Chemical Society, vol.74, p.676 (1952); and Journal of the American Chemical Society, vol.86, p.1839 (1964)].

[0028] [Preparation Process (B)]

[0029] [Chem. Formula 3]

Formula [4] Formula [5]



Formula [1]

[0030] (wherein R and X denote the same as defined above, and Z denotes a halogen atom or the ROC(O)O– group).

[0031] An amino acid amide derivative represented by the formula [1] can be prepared by the reaction of a compound represented by the formula [4] with amines represented by the formula [5] or inorganic acid salts such as hydrochlorides, etc. or organic acid salts such as tosylate salts, etc. with or without the presence of a base.

[0032] A solvent usable in the reaction may be any solvent as far as it does not inhibit the reaction, and its examples may include hydrocarbons such as pentene, hexane, heptane, cyclohexane, petroleum ether, ligroin, benzene, toluene, xylene, etc., halogenated hydrocarbons such as dichloromethane, dichloroethane, chloroform, carbon tetrachloride, chlorobenzene, dichlorobenzene, etc., ethers such as diethylether, diisopropylether, ethyleneglycol dimethylether, tetrahydrofuran, dioxane, etc., ketones such as acetone, methylethylketone, methylisopropylketone,

methylisobutylketone, etc., acetic esters such as methyl acetate, ethyl acetate, etc., nitriles such as acetonitrile, propionitrile, benzonitrile, etc., aprotic polar solvents such as dimethylsulfolane, dimethylformamide, sulfolane, etc. and solvent mixtures combining members selected from these solvents.

[0033] A base generally used in such a type of reaction can be used. Its examples may include alkali metal hydroxides such as sodium hydroxide, potassium hydroxide, etc., alkali earth metal hydroxides such as calcium hydroxide, etc., alkali metal carbonates such as sodium carbonate, potassium carbonate, etc., organic bases such as triethyleneamine, trimethyleneamine, dimethylaniline, pyridine, N-methylpyridine, 1,5-diazabicyclo[4,3,0]non-5-ene (DBN), 1,8-dizabicyclo-[5,4,0]-undecene-7-ene (DBU), etc. Preferable examples are tertiary amines such as triethyleneamine, pyridine, N-methylpyridine, etc. A reaction temperature is within the range of –20°C~100°C, preferably within the range of 0°C~40°C. A reaction time is preferably 30 minutes~20 hours.

[0034] Now, the preparation of compounds to be used according to the present invention shall be specifically explained in reference to referential preparation examples.

[0035] [Referential Preparation Example 1: Preparation of N^2-tert-butoxycarbonyl-N^1-[(R)-1-(6-chloror-2-benzothiazolyl)ethyl]-L-valine amide (Compound No.5)]

In 50mL of dichloromethane, 0.8g of N-tert-butoxycarbonyl-L-valine was dissolved, and with the addition of 0.37g of N-methylpiperidine at –20°C, the mixture was stirred at the temperature for 10 minutes. Then, after the addition of 0.8g of (R)-1-(6-chloro-2-benzothiazolyl)ethylamine to this mixture, a coolant was removed and at room temperature, stirring was

continued for 15 hours. Water was added to this reaction mixture, the dichloromethane layer was washed with an aqueous 5% sodium hydrogen carbonate solution and then with water, to be followed by desiccation with anhydrous magnesium sulfate and solvent removal by distillation under vacuum. Crude crystals obtained as residues were chromatographically purified using a silica gel column to obtain 1.3g of the intended substance as colorless prismatic crystals (at the yield of 87%).

[0036] [Referential Preparation Example 2: Preparation of N^1-[(R)-1-(6-fluoro-2-benzothiazolyl)ethyl]- N^2-isopropoxycarbonyl-L- valine amide (Compound No.10)]

In 25mL of dichloromethane, 0.8g of N-isopropoxycarbonyl-L-valine was dissolved, and with the addition of 0.4g of N-methylpiperidine at –20°C, the mixture was stirred at the temperature for 10 minutes. Then, after the addition of 0.6g of isobutyl chloroformate, the mixture was stirred for an hour at a temperature of –20°C~–10°C. After the addition of 0.8g of (R)-1-(6-fluoro-2-benzothiazolyl)ethylamine to this mixture at –60°C, a coolant was removed and stirring was continued until room temperature was reached. Water was added to this reaction mixture, the dichloromethane layer was washed with an aqueous 5% sodium hydrogen carbonate solution and then with water, to be followed by desiccation with anhydrous magnesium sulfate and distillation under vacuum. Crude crystals obtained were chromatographically purified using a silica gel column to obtain 0.95g of the intended substance as white powder (at the yield of 63%).

[0037]

[Mode of Working Invention] A fungicide composition according to the present invention is to be used as a preparation generally used in the

pesticide preparation such as powder agents, granular agents, fine granular agents, tablet agents, liquid agents, wettable agents, flowable agents, aerosol agents, etc. These preparations can be used according to general application methods such as seed treatment, application to leaves and stems, soil treatment, water pad application, etc.

[0038] The application amount of a fungicide composition according to the present invention is variable depending upon the kind of fungicidally active compounds to be combined, disease injuries, as the control object, infection trends, injury levels, environmental conditions, types of the preparations to be used, etc. A blending weight ratio of an amino acid amide derivative represented by the formula [1] and one or more fungicidal compounds selected from the group of (A) to (X) ranges from 1:0.5 to 1:1000, and the preparation is generally applied at a rate of 1g~5kg per are.

[0039] The present invention shall be explained in more detail. When an amino acid amide derivative represented by the formula [1] is combined with a fungicidal compound having no therapeutic effects for downy mildew of cucumbers, downy mildew of grapes, late blight of tomatoes, etc., such as (A) zinc ion-coordinated manganese ethylene- bisdithiocarbamate (Manzeb), (B) manganese ethylenebisdithiocarbamate (Maneb), (C) zinc ethylenebisdithiocarbamate, (Zineb), (D) zinc propylene-bisdithiocarbamate (Propineb), (E) N-(trichloromethylthio)cyclohexa-4-en-1,2-dicaroboxyimide (Captan), (F) tetrachlorophthalonitrile (TPN), (G) 3-chloro-N-(3-chloro- 2,6-dinitro-4- α, α, α-trifluoromethyl-2-pyridine) amine (Flurazinam), (H) 2,3-dichloro- 4-fluorophenylmaleimide (Fluoroimide), etc. or when the combination is with a fungicidal compound without sufficient durability of the agent for controlling Phycomycetes such as (I) 1-(2-cyano-2-methoxyiminoacetyl)-3-ethylurea, (Q) (E,Z)-4-[3-

(4-chlorophenyl)-3-(3,4-dimethoxyphenyl) acryloyl]morpholine, etc., a blending ratio (weight ratio) of the fungicidal compound for agricultural and horticultural uses based on a part of the amino acid amine derivative represented y the formula [1] is within a range of 1~1000 parts, preferably 1~100 parts. When these fungicide compositions for agricultural and horticultural uses according to the present invention are used as powder agents and granular agents, the preparations are to be applied at a ratio of 1g~5kg, preferably 1g~1kg, per 10 ares as the active components. When those are used as emulsion agents or wettable agents, the preparations are applied upon the dilution to a concentration range of 1~5,000ppm, preferably 10~2,000ppm.

[0040] When an amino acid amide derivative represented by the formula [1] is combined with a fungicidal compound having both the preventive and therapeutic effects for downy mildew of cucumbers, downy mildew of grapes, late blight of tomatoes, etc., such as (M) methyl-N-(2-methoxy-acetyl)- N-(2,6-xylyl)-DL-alaninate (Metraxyl), (N) 2-methoxy-N-(2-oxo-1,3-oxazolidine-3-il)acetamide-2',6'-xylidine(Oxadixyl), (O) methyl-N-phenylacetyl-N-2,6-xylyl-DL-alaninate (Benalaxyl), (P) methyl-N-(2-furyl)-N-(2,6-xylyl)- DL-alaninate (Furalaxyl), etc., a blending ratio (weight ratio) of the fungicidal compound for agricultural and horticultural uses based on a part of the amino acid amine derivative represented by the formula [1] is within a range of 0.5~100 parts, preferably 0.5~50 parts. When these fungicide compositions for agricultural and horticultural uses according to the present invention are used as powder agents and granular agents, the preparations are to be applied at a ratio of 1g~2kg as the active components, preferably 10g~500g, per 10 ares. When those are used as

emulsion agents or wettable agents, the preparations are applied upon the dilution to a concentration range of 1~5,000ppm, preferably 10~1,000ppm.

[0041] When an amino acid amide derivative represented by the formula [1] is combined with a chemical agent and [sic] a fungicidal compound having no activity to downy mildew of cucumbers, downy mildew of grapes, late blight of tomatoes, etc., such as (T) (RS)-2-(2,4-dichlorophenyl)-1-(1H-1,2,4-triazol-2-il)- hexane-2-ol (Hexaconazol), (U) (cis, trans)-3-chloro-4-[4-methyl-2-(1H-1,2,4-triazol-1-ilmethyl)-1,3-dioxolane-2-il]phenyl-4-chloro phenylether, (Difenaconazol), (V) (E)-4-chloro-α, α, α-trifluoro-N-(1-imidazol-1-il- 2-propoxyethylidene)-o-toluidine (Triflumizol), (W) 2,4'-chloro-α- (pyrimidine-5-il)-benzhydrylalcohol (Fenarimol), and (X) 1,1'-iminodi- (octamethylene)diguanidium tris(dodecylbenzenesulfonate) (iminoctazine dodecylbenzensulfonate), etc., a blending ratio (weight ratio) of the fungicidal compound for agricultural and horticultural uses based on a part of the amino acid amine derivative represented by the formula [1] is within a range of 1~1000 parts, preferably 1~500 parts. When these fungicide compositions for agricultural and horticultural uses according to the present invention are used as powder agents and granular agents, the preparations are to be applied at a ratio of 1g~5kg, preferably 1g~1kg, per 10 ares, as the active components. When those are used as emulsion agents or wettable agents, the preparations are applied upon the dilution with water to a concentration range of 1~5,000ppm, preferably 10~2,000ppm.

[0042] A fungicide composition for agricultural and horticultural uses according to the present invention may be prepared by blending an amino acid amide derivative with 2 or more kinds of fungicidal compounds for agricultural and horticultural uses. A typical example may be 3-component mixtures comprising an amino acid amide derivative and two fungicidal

compounds for agricultural and horticultural uses: such as (A) and (I), (A) and (J), (R) and (I), and (R) and (J).

[0043] A fungicide composition for agricultural and horticultural uses according to the present invention is preferably to be applied as preparations such as granular agents, wettable agents, emulsions agents, finely granular agents, powder agents, flowable agents, etc. formed according to conventional methods by formulating with carriers, surfactants, dispersing agents, supplemental additives, etc. in addition to said active components. Examples of a preferable carrier may include solid carriers such as talc, bentonite, clay, kaolin, diatomaceous earth, calcium carbonate, white carbon, vermiculite, slaked lime, silica sand, ammonium sulfate, urea, etc., liquid carriers such as isopropylalcohol, xylene, cyclohexanone, methylnaphthalene, fatty acid esters, vegetable oils, mineral oils, animal oils, water, etc. and the like. Examples of a surfactant and a dispersing agent may include polyoxyethylene sorbitan fatty acid esters, polyoxyethylene fatty acid esters, polyoxyethylene castor oil, polyoxyethylene alkylethers, polyoxyethylene alkylphenylethers, polyoxyethylene alkylether sulfate, alkylbenzene sulfonate, naphthalenesulfonate-formalin condensation products, lignin sulfonate, polyoxyethylene alkylphenylether sulfonate, etc. Examples of a supplemental additive may include carboxymethylcellulose, polyvinyl alcohol, polyethylene glycol, propylene glycol, gum Arabic, dextrin, condensation phosphates, etc. These preparations are applied either upon dilution to an appropriate concentration or directly as it is.

[0044] Furthermore, a fungicide composition for agricultural and horticultural uses according to the present invention may be optionally

blended as required with insecticides, other fungicides, herbicides, plant growth regulators, fertilizers, etc.

[0045] [Examples]

Preparatory Example 1: Wettable agent.

A wettable agent was prepared by homogeneously pulverizing 1% of a compound (3), 5% of a fungicidal compound (A) for agricultural and horticultural uses, 1.5% of a naphthalene sulfonate-formaldehyde condensate ion product sodium salt, 1.5% of polyoxyethylene alkylaryl, 26% of diatomaceous earth and 65% of clay.

[0046] Preparatory Example 2: Wettable agent.

A wettable agent was prepared by homogeneously pulverizing 1% of a compound (3), 10% of a fungicidal compound (B) for agricultural and horticultural uses, 1.5% of a naphthalene sulfonate-formaldehyde condensate ion product sodium salt, 1.5% of polyoxyethylene alkylaryl, 26% of diatomaceous earth and 60% of clay.

[0047] Preparatory Example 3: Wettable agent.

A wettable agent was prepared by homogeneously pulverizing 1% of a compound (8), 15% of a fungicidal compound (E) for agricultural and horticultural uses, 1.5% of a naphthalene sulfonate-formaldehyde condensate ion product sodium salt, 1.5% of polyoxyethylene alkylaryl, 26% of diatomaceous earth and 55% of clay.

[0048] Preparatory Example 4: Wettable agent.

A wettable agent was prepared by homogeneously pulverizing 1% of a compound (3), 20% of a fungicidal compound (A) for agricultural and horticultural uses, 1.5% of a naphthalene sulfonate-formaldehyde condensate ion product sodium salt, 1.5% of polyoxyethylene alkylaryl, 26% of diatomaceous earth and 50% of clay.

[0049] Preparatory Example 5: Wettable agent.

A wettable agent was prepared by homogeneously pulverizing 0.1% of a compound (7), 3% of a fungicidal compound (H) for agricultural and horticultural uses, 1.5% of a naphthalene sulfonate-formaldehyde condensate ion product sodium salt, 1.5% of polyoxyethylene alkylaryl, 26% of diatomaceous earth and 67.9% of clay.

[0050] Preparatory Example 6: Wettable agent.

A wettable agent was prepared by homogeneously pulverizing 0.1% of a compound (7), 4% of a fungicidal compound (A) for agricultural and horticultural uses, 1.5% of a naphthalene sulfonate-formaldehyde condensate ion product sodium salt, 1.5% of polyoxyethylene alkylaryl, 26% of diatomaceous earth and 66.9% of clay.

[0051] Preparatory Example 7: Wettable agent.

A wettable agent was prepared by homogeneously pulverizing 0.1% of a compound (8), 4% of a fungicidal compound (B) for agricultural and horticultural uses, 1.5% of a naphthalene sulfonate-formaldehyde condensate ion product sodium salt, 1.5% of polyoxyethylene alkylaryl, 26% of diatomaceous earth and 65.9% of clay.

[0052] Preparatory Example 8: Wettable agent.

A wettable agent was prepared by homogeneously pulverizing 0.1% of a compound (3), 10% of a fungicidal compound (D) for agricultural and horticultural uses, 1.5% of a naphthalene sulfonate-formaldehyde condensate ion product sodium salt, 1.5% of polyoxyethylene alkylaryl, 26% of diatomaceous earth and 60.9% of clay.

[0053] Preparatory Example 9: Wettable agent.

A wettable agent was prepared by homogeneously pulverizing 0.1% of a compound (10), 15% of a fungicidal compound (A) for agricultural and

horticultural uses, 1.5% of a naphthalene sulfonate-formaldehyde condensate ion product sodium salt, 1.5% of polyoxyethylene alkylaryl, 26% of diatomaceous earth and 55.9% of clay.

[0054] Preparatory Example 10: Wettable agent.

A wettable agent was prepared by homogeneously pulverizing 0.1% of a compound (7), 20% of a fungicidal compound (C) for agricultural and horticultural uses, 1.5% of a naphthalene sulfonate-formaldehyde condensate ion product sodium salt, 1.5% of polyoxyethylene alkylaryl, 26% of diatomaceous earth and 50.9% of clay.

[0055] Preparatory Example 11: Wettable agent.

A wettable agent was prepared by homogeneously pulverizing 3% of a compound (3), 5% of a fungicidal compound (A) for agricultural and horticultural uses, 1.5% of a naphthalene sulfonate-formaldehyde condensate ion product sodium salt, 1.5% of polyoxyethylene alkylaryl, 26% of diatomaceous earth and 50.9% of clay.

[0056] Preparatory Example 12: Wettable agent.

A wettable agent was prepared by homogeneously pulverizing 0.3% of a compound (7), 5% of a fungicidal compound (A) for agricultural and horticultural uses, 1.5% of a naphthalene sulfonate-formaldehyde condensate ion product sodium salt, 1.5% of polyoxyethylene alkylaryl, 26% of diatomaceous earth and 65.7% of clay.

[0057] Preparatory Example 13: Wettable agent.

A wettable agent was prepared by homogeneously pulverizing 0.3% of a compound (3), 10% of a fungicidal compound (A) for agricultural and horticultural uses, 1.5% of a naphthalene sulfonate-formaldehyde condensate ion product sodium salt, 1.5% of polyoxyethylene alkylaryl, 26% of diatomaceous earth and 60.7% of clay.

[0058] Preparatory Example 13: Emulsified agent.

An emulsified agent was prepared by homogeneously dissolving 1% of a compound (3), 20% of a fungicidal compound (A) for agricultural and horticultural uses, 25% of cyclohexanone, 13% of polyoxyethylene alkylaryl ether, 4% of calcium alkylbenzene sulfonate and 37% of methylnaphthalene.

[0059] Preparatory Example 14: Powder agent.

A powder agent was prepared by blending and pulverizing 0.3% of a compound (8), 0.5% of a fungicidal compound (A) for agricultural and horticultural uses, 6.2% of diatomaceous earth and 93% of clay.

[0060] Preparatory Example 16: Granular agent.

A granular agent was prepared by homogeneously blending and pulverizing 0.3% of a compound (10), 5% of a fungicidal compound (A) for agricultural and horticultural uses, 2% of sodium lauryl sulfonate, 4.7% of sodium lignin sulfonate, 2% of carboxymethylcellulsoe and 86% of clay, kneading a mixture comprising 100 parts by weight of the pulverized mixture with 20 parts by weight of added water, processing the kneaded material to 14~32 mesh sized granules with an extruder-type granulator, and drying the granules.

[0061] Preparatory Example 17: Flowable agent.

A flowable agent was prepared by blending 1% of a compound (10), 20% of a fungicidal compound (A) for agricultural and horticultural uses, 6% of a sodium ligninsulfonate, 1% of sodium polyoxyethylene alkylnaphthalene sulfonate and 0.1% of xanthan gum with the addition of 71.9% of water.

[0062] Preparatory Example 18: Wettable agent.

A wettable agent was prepared by homogeneously blending and pulverizing 1% of a compound (5), 1% of a fungicidal compound (M) for

agricultural and horticultural uses, 1.5% of a naphthalene sulfonate-formaldehyde condensation product sodium salt, 1.5% of polyoxyethylene alkylaryl, 26% of diatomaceous earth and 69% of clay.

[0063] Preparatory Example 19: Wettable agent.

A wettable agent was prepared by homogeneously pulverizing 1% of a compound (3), 5% of a fungicidal compound (A) for agricultural and horticultural uses, 1.5% of a naphthalene sulfonate-formaldehyde condensate ion product sodium salt, 1.5% of polyoxyethylene alkylaryl, 26% of diatomaceous earth and 65% of clay.

[0064] Preparatory Example 20: Wettable agent.

A wettable agent was prepared by homogeneously pulverizing 1% of a compound (3), 5% of a fungicidal compound (A) for agricultural and horticultural uses, 1.5% of a naphthalene sulfonate-formaldehyde condensate ion product sodium salt, 1.5% of polyoxyethylene alkylaryl, 26% of diatomaceous earth and 65% of clay.

[0065] Preparatory Example 21: Wettable agent.

A wettable agent was prepared by homogeneously pulverizing 1% of a compound (7), 4% of a fungicidal compound (Q) for agricultural and horticultural uses, 1.5% of a naphthalene sulfonate-formaldehyde condensate ion product sodium salt, 1.5% of polyoxyethylene alkylaryl, 26% of diatomaceous earth and 66% of clay.

[0066] Preparatory Example 22: Wettable agent.

A wettable agent was prepared by homogeneously pulverizing 1% of a compound (3), 5% of a fungicidal compound (M) for agricultural and horticultural uses, 1.5% of a naphthalene sulfonate-formaldehyde condensate ion product sodium salt, 1.5% of polyoxyethylene alkylaryl, 26% of diatomaceous earth and 65% of clay.

[0067] Preparatory Example 23: Wettable agent.

A wettable agent was prepared by homogeneously pulverizing 1% of a compound (7), 6% of a fungicidal compound (Q) for agricultural and horticultural uses, 1.5% of a naphthalene sulfonate-formaldehyde condensate ion product sodium salt, 1.5% of polyoxyethylene alkylaryl, 26% of diatomaceous earth and 64% of clay.

[0068] Preparatory Example 24: Wettable agent.

A wettable agent was prepared by homogeneously pulverizing 1% of a compound (5), 10% of a fungicidal compound (M) for agricultural and horticultural uses, 1.5% of a naphthalene sulfonate-formaldehyde condensate ion product sodium salt, 1.5% of polyoxyethylene alkylaryl, 26% of diatomaceous earth and 64% of clay.

[0069] Preparatory Example 25: Wettable agent.

A wettable agent was prepared by homogeneously pulverizing 1% of a compound (3), 15% of a fungicidal compound (I) for agricultural and horticultural uses, 1.5% of a naphthalene sulfonate-formaldehyde condensate ion product sodium salt, 1.5% of polyoxyethylene alkylaryl, 26% of diatomaceous earth and 55% of clay.

[0070] Preparatory Example 26: Wettable agent.

A wettable agent was prepared by homogeneously pulverizing 1% of a compound (10), 20% of a fungicidal compound (I) for agricultural and horticultural uses, 1.5% of a naphthalene sulfonate-formaldehyde condensate ion product sodium salt, 1.5% of polyoxyethylene alkylaryl, 26% of diatomaceous earth and 50% of clay.

[0071] Preparatory Example 27: Wettable agent.

A wettable agent was prepared by homogeneously pulverizing 0.1% of a compound (3), 3% of a fungicidal compound (M) for agricultural and

horticultural uses, 1.5% of a naphthalene sulfonate-formaldehyde condensate ion product sodium salt, 1.5% of polyoxyethylene alkylaryl, 26% of diatomaceous earth and 67.9% of clay.

[0072] Preparatory Example 28: Wettable agent.

A wettable agent was prepared by homogeneously pulverizing 0.1% of a compound (10), 5% of a fungicidal compound (M) for agricultural and horticultural uses, 1.5% of a naphthalene sulfonate-formaldehyde condensate ion product sodium salt, 1.5% of polyoxyethylene alkylaryl, 26% of diatomaceous earth and 67.9% of clay.

[0073] Preparatory Example 29: Wettable agent.

A wettable agent was prepared by homogeneously pulverizing 0.1% of a compound (3), 10% of a fungicidal compound (P) for agricultural and horticultural uses, 1.5% of a naphthalene sulfonate-formaldehyde condensate ion product sodium salt, 1.5% of polyoxyethylene alkylaryl, 26% of diatomaceous earth and 60.9% of clay.

[0074] Preparatory Example 30: Wettable agent.

A wettable agent was prepared by homogeneously pulverizing 3% of a compound (10), 5% of a fungicidal compound (M) for agricultural and horticultural uses, 1.5% of a naphthalene sulfonate-formaldehyde condensate ion product sodium salt, 1.5% of polyoxyethylene alkylaryl, 26% of diatomaceous earth and 63% of clay.

[0075] Preparatory Example 31: Wettable agent.

A wettable agent was prepared by homogeneously pulverizing 0.3% of a compound (9), 5% of a fungicidal compound (M) for agricultural and horticultural uses, 1.5% of a naphthalene sulfonate-formaldehyde condensate ion product sodium salt, 1.5% of polyoxyethylene alkylaryl, 26% of diatomaceous earth and 65.7% of clay.

[0076] Preparatory Example 32: Wettable agent.

A wettable agent was prepared by homogeneously pulverizing 0.3% of a compound (3), 10% of a fungicidal compound (I) for agricultural and horticultural uses, 1.5% of a naphthalene sulfonate-formaldehyde condensate ion product sodium salt, 1.5% of polyoxyethylene alkylaryl, 26% of diatomaceous earth and 60.7% of clay.

[0077] Preparatory Example 33: Wettable agent.

A wettable agent was prepared by homogeneously pulverizing 2% of a compound (11), 1% of a fungicidal compound (M) for agricultural and horticultural uses, 1.5% of a naphthalene sulfonate-formaldehyde condensate ion product sodium salt, 1.5% of polyoxyethylene alkylaryl, 26% of diatomaceous earth and 68% of clay.

[0078] Preparatory Example 34: Emulsified agent.

An emulsified agent was prepared by homogeneously dissolving 1% of a compound (5), 20% of a fungicidal compound (M) for agricultural and horticultural uses, 25% of cyclohexanone, 13% of polyoxyethylene alkylaryl ether, 4% of calcium alkylbenzene sulfonate and 37% of methylnaphthalene.

[0079] Preparatory Example 35: Powder agent.

A powder agent was prepared by blending and pulverizing 0.3% of a compound (8), 0.5% of a fungicidal compound (M) for agricultural and horticultural uses, 6.2% of diatomaceous earth and 93% of clay.

[0080] Preparatory Example 36: Granular agent.

A granular agent was prepared by homogeneously blending and pulverizing 0.3% of a compound (10), 5% of a fungicidal compound (M) for agricultural and horticultural uses, 2% of sodium lauryl sulfonate, 4.7% of sodium lignin sulfonate, 2% of carboxymethylcellulsoe and 86% of clay,

kneading a mixture comprising 100 parts by weight of the pulverized mixture and 20 parts by weight of added water, processing the kneaded material to 14~32 mesh sized granules with an extruder-type granulator, and drying the granules.

[0081] Preparatory Example 37: Flowable agent.

A flowable agent was prepared by blending and pulverizing 1% of a compound (10), 10% of a fungicidal compound (M) for agricultural and horticultural uses, 4% of ammonium polyoxyethylene nonylphenylether, 1.5% of sodium polyoxyethylene naphthalene sulfonate and 0.15% of xanthan gum with the addition of 83.35% of water.

[0082] Preparatory Example 38: Emulsified agent.

An emulsified agent was prepared by homogeneously dissolving 1% of a compound (5), 20% of a fungicidal compound (M) for agricultural and horticultural uses, 25% of cyclohexanone, 13% of polyoxyethylene alkylaryl ether, 4% of calcium alkylbenzene sulfonate and 37% of methylnaphthalene.

[0083] Preparatory Example 39: Powder agent.

A powder agent was prepared by blending and pulverizing 0.3% of a compound (8), 0.5% of a fungicidal compound (M) for agricultural and horticultural uses, 6.2% of diatomaceous earth and 93% of clay.

[0084] Preparatory Example 40: Granular agent.

A granular agent was prepared by homogeneously blending and pulverizing 0.3% of a compound (10), 5% of a fungicidal compound (M) for agricultural and horticultural uses, 2% of sodium lauryl sulfonate, 4.7% of sodium lignin sulfonate, 2% of carboxymethylcellulsoe and 86% of clay, kneading a mixture comprising 100 parts by weight of the pulverized mixture and 20 parts by weight of added water, processing the kneaded

material to 14~32 mesh sized granules with an extruder-type granulator, and drying the granules.

[0085] Preparatory Example 41: Wettable agent.

A wettable agent was prepared by homogeneously pulverizing 2% of a compound (3), 5% of a fungicidal compound (T) for agricultural and horticultural uses, 1.5% of a naphthalene sulfonate-formaldehyde condensation product sodium salt, 1.5% of polyoxyethylene alkylaryl, 26% of diatomaceous earth and 65.9% of clay.

[0086] Preparatory Example 42: Wettable agent.

A wettable agent was prepared by homogeneously pulverizing 0.1% of a compound (10), 10% of a fungicidal compound (X) for agricultural and horticultural uses, 1.5% of a naphthalene sulfonate-formaldehyde condensate ion product sodium salt, 1.5% of polyoxyethylene alkylaryl, 26% of diatomaceous earth and 60.9% of clay.

[0087] Preparatory Example 43: Wettable agent.

A wettable agent was prepared by homogeneously pulverizing 0.3% of a compound (8), 50% of a fungicidal compound (U) for agricultural and horticultural uses, 1.5% of a naphthalene sulfonate-formaldehyde condensate ion product sodium salt, 1.5% of polyoxyethylene alkylaryl, 26% of diatomaceous earth and 20.7% of clay.

[0088] Preparatory Example 44: Emulsified agent.

An emulsified agent was prepared by homogeneously dissolving 1% of a compound (5), 20% of a fungicidal compound (T) for agricultural and horticultural uses, 25% of cyclohexanone, 13% of a polyoxyethylene alkylarylether, 4% of calcium alkylbenzene sulfonate and 37% of methylnaphthalene.

[0089] Preparatory Example 45: Powder agent.

A powder agent was prepared by blending and pulverizing 0.3% of a compound (8), 0.5% of a fungicidal compound (T) for agricultural and horticultural uses, 6.2% of diatomaceous earth and 93% of clay.

[0090] Preparatory Example 46: Granular agent.

A granular agent was prepared by homogeneously blending and pulverizing 0.3% of a compound (10), 5% of a fungicidal compound (T) for agricultural and horticultural uses, 2% of sodium lauryl sulfonate, 4.7% of sodium lignin sulfonate, 2% of carboxymethylcellulsoe and 86% of clay, kneading a mixture comprising 100 parts by weight of the pulverized mixture with 20 parts by weight of added water, processing the kneaded material to 14~32 mesh sized granules with an extruder-type granulator, and drying the granules.

[0091] Preparatory Example 47: Flowable agent.

A flowable agent was prepared by blending 1% of a compound (10), 20% of a fungicidal compound (T) for agricultural and horticultural uses, 6% of ammonium polyoxyethylene nonylphenylether, and 0.15% of xanthan gum with the addition of 72.85% of water.

[0092] Next, the fungicidal effect of a fungicide composition for agricultural and horticultural uses according to the present invention shall be explained in detail with reference to test examples.

[0093] Test Example 1: Testing the effect of protecting cucumbers from downy mildew.

Ten (10) cucumber seeds (the kind: Sagami Hampaku) were sown in every 9cm×9cm sized PVC-made pot for 7-day growth in a greenhouse chamber, and cucumber seedlings having developed cotyledons were used as test plants. Compositions according to the present invention, as prepared after the preparatory examples 1, 2, 5, 7, 8, 18, 19, 21, 25, 27, 28,

29, 33, 41 and 42 were diluted with water to predetermined concentrations for respectively spraying at the ratio of 15mL a pot. After air drying, the test plants were inoculated with a conidia spore suspension of Pseudoperonospora cubensis by spraying, and then were immediately placed in a moist chamber kept at 20C, for 24 hours. Later, the test plants were moved into a greenhouse chamber, and the infection level of each cotyledon was examined according to the below described infection index. Based on the observed index values, levels of injuries were derived, and control factors were further derived after the numeric equation 2.

[0094]

Infection index 0: No infection observed.

1: Infected area less than 5%

2: Infected area more than 5% but less than 33.3%

3: Infected area more than 33.3% but less than 66.6%

4: Infected area more than 66.6% or defoliation.

[0095] [Numeric equation 1]

$$InjuryLevel(\%) = \frac{\sum(Index \times NumberofLeaves) \times 100}{NumberofExa\,\min edLeaves}$$

[0096] [Numeric equation 2]

$$ControlFactor = \left(1 - \frac{InjuryLevelinTreatedBlock}{InjuryLevelinNon-TreatedBlock}\right) \times 100$$

[0097] [Table 2] (cx = comparative example)

test compound			3+A	3+B	3+C	3+F	3+G	3+I	3+L
active component conc, ppm			1+5	1+5	1+50	1+5	1+5	1+15	1+50
control factor, %			71	58	56	80	74	62	73
3+M	3+Q	3+T	3+U	3+X	5+A	5+B	5+D	5+E	5+H
1+0.5	1+2	1+50	1+50	1+50	0.5+0.5	0.5+0.5	0.5+0.5	0.5+15	0.5+15
76	68	42	39	50	75	71	65	58	62
5+I	5+J	5+K	5+L	5+M	5+Q	5+V	5+W	7+A	7+B
0.5+15	0.5+60	0.5+50	0.5+50	0.5+0.5	0.5+2	0.5+50	0.5+50	0.5+5	0.5+5
64	77	76	64	78	75	51	54	82	76

(continued)

test compound			7+C	7+D	7+E	7+F	7+G	7+H	7+I
active component conc, ppm			0.5+50	0.5+5	0.5+15	0.5+5	0.5+5	0.5+15	0.5+15
control factor, %			75	80	68	91	84	79	82
7+J	7+K	7+L	7+M	7+Q	7+U	7+V	7+W	8+A	8+B
0.5+50	0.5+50	0.5+50	0.5+0.5	0.5+2	0.5+50	0.5+50	0.5+50	0.5+5	0.5+5
84	86	64	88	79	60	63	58	84	73
8+D	8+I	8+J	8+K	8+Q	8+V	8+W	10+A	10+B	10+C
0.5+5	0.5+15	0.5+50	0.5+50	0.5+2	0.5+50	0.5+50	0.5+5	0.5+5	0.5+50
77	74	81	80	83	57	62	87	74	81
10+D	10+E	10+F	10+G	10+H	10+I	10+J	10+K	10+L	10+M
0.5+5	0.5+15	0.5+5	0.5+5	0.5+15	0.5+15	0.5+50	0.5+50	0.5+50	0.5+0.5
81	73	86	85	80	73	86	88	69	85
10+Q	10+T	10+U	12+A	12+B	12+D	12+M	12+T	12+W	12+X
0.5+2	0.5+50	0.5+50	1+5	1+5	1+50	1+0.5	1+50	1+50	1+50
84	59	62	91	82	88	87	69	64	67
3 cx	5cx	7cx	8cx	10cx	11cx	A cx	B cx	C cx	D cx
1	0.5	0.5	0.5	0.5	1	5	5	50	5
24	32	44	40	45	53	46	28	32	37
E cx	F cx	G cx	H cx	I cx	J cx	K cx	L cx	M cx	Q cx
15	5	5	15	15	50	50	50	0.5	2
12	51	43	31	28	41	43	24	46	39
T cx	U cx	V cx	W cx	X cx					
50	50	50	50	50					
0	0	0	0	0					

[0098] Test Example 2: Testing the effect of protecting tomatoes from late blight.

Tomato seedlings (the kind: Ponte Rosa) were transplanted one by one in 12cm diameter unglazed pots for growth in a greenhouse chamber, and tomato seedlings developed to have 6~7 compound leaves were used as test plants. Compositions according to the present invention, as prepared after the preparatory examples 2, 4, 6, 7, 8, 10, 20, 22, 23, 24, 26, 41 and 42 were diluted with water to contain respective active compounds or active compound compositions in predetermined concentrations of active components for respectively spraying 20mL a pot. After air drying, test plants were inoculated with a swarm ascus suspension of Phytophthora infestans by spraying, and then, were immediately placed in a moist chamber kept at 20C. Four (4) days later, the infection level of each leaflet was examined according to the infection index described in Example 1, and based on the observed index values, levels of injuries and control factors

were derived in the same manner as in Example 1. Test results are shown in Table 3.

[0099] [Table 3]

test compound			3+A	3+B	3+D	3+F	3+G	3+I	3+M
active component conc, ppm			1+20	1+50	1+100	1+10	1+10	1+10	1+5
control factor, %			72	70	68	70	76	67	76
3+Q	3+U	3+X	5+A	5+B	5+C	5+D	5+G	5+I	5+M
1+3	1+50	1+50	0.5+20	0.5+50	0.5+100	0.5+100	0.5+10	0.5+10	0.5+5
79	55	48	71	78	69	71	79	72	80
5+Q	5+T	5+V	5+W	7+A	7+B	7+C	7+D	7+F	7+G
0.5+3	0.5+50	0.5+50	0.5+50	0.5+20	0.5+50	0.5+100	0.5+100	0.5+10	0.5+10
81	54	49	53	75	77	67	72	75	79
7+I	7+M	7+Q	7+T	7+U	8+A	8+B	8+C	8+D	8+F
0.5+10	0.5+5	0.5+3	0.5+50	0.5+50	0.5+20	0.5+50	0.5+100	0.5+100	0.5+10
71	74	80	56	53	82	80	73	79	84
8+G	8+I	8+M	8+Q	8+U	8+V	10+A	10+B	10+C	10+D
0.5+10	0.5+10	0.5+5	0.5+3	0.5+50	0.5+50	0.5+20	0.5+50	0.5+100	0.5+10
78	75	84	82	58	60	82	89	74	78
10+F	10+G	10+I	10+M	10+Q	10+T	10+X	11+A	11+B	11+F
0.5+10	0.5+10	0.5+10	0.5+5	0.5+3	0.5+50	0.5+50	1+20	1+50	1+10
81	92	85	90	87	64	61	73	71	79
11+I	11+M	11+Q	11+U	11+V	11+X	3 cx	5 cx	7 cx	8 cx
1+10	1+5	1+3	1+50	1+50	1+50	1	0.5	0.5	0.5
76	82	88	54	49	57	32	31	37	41
10 cx	11 cx	A cx	B cx	C cx	D cx	F cx	G cx	I cx	M cx
0.5	1	20	50	100	100	10	10	10	5
47	36	32	35	17	29	34	42	28	37
Q cx	T cx	U cx	V cx	W cx	X cx				
3	50	50	50	50	50				
42	0	0	0	0	0				

[0100] Test Example 3: Testing the effect of protecting grapes from downy mildew.

Grape nursery stocks (the kind: Kyoho) planted in 12cm diameter unglazed pots were pruned and grown in a greenhouse chamber, and the grape nursery stocks developed to have 4~5 leaves were used as test plants. Compositions according to the present invention, as prepared after the preparatory examples 1, 2, 3, 12, 20, 23, 24, 29, 30, 32, 33, 41 and 42 were diluted with water to contain respective active compounds or active compound compositions in predetermined concentrations of active components for respectively spraying 20mL a pot. After air drying, test plants were inoculated with a conidia spore suspension of Plasmopara viticola by spraying, then, immediately placed in a moist chamber kept at

20°C for 24 hours, and later, moved in a greenhouse chamber kept at 20C for 24 hours for pathogenic development. Ten (10) days later, the test plants were again placed in a moist chamber kept at 20°C for 24 hours so that conidia spores were formed. The size of infected areas including leaf positions where conidia spores were formed and lesion positions with brown spots and brown pustules were examined according to the standard of infection index described in Example 1, and, based on the observed index values, levels of injuries and control factors were derived in the same manner as in Example 1. Test results are shown in Table 4.

[0101] [Table 4]

test compound			3+A	3+F	3+I	3+M	3+T	3+W	5+F
active component conc, ppm			1+5	1+5	1+10	1+0.5	1+50	1+50	0.5+5
control factor, %			77	81	84	84	58	55	83
5+Q	5+U	5+V	7+A	7+F	7+I	7+M	7+Q	7+T	7+U
0.5+3	0.5+50	0.5+50	0.3+5	0.3+5	0.3+10	0.3+0.5	0.3+3	0.3+3	0.3+50
82	56	52	73	77	74	82	84	52	51
8+A	8+F	8+M	8+Q	8+T	8+U	10+A	10+F	10+I	10+M
0.3+5	0.3+5	0.3+0.5	0.3+3	0.3+50	0.3+50	0.3+5	0.3+5	0.3+10	0.3+0.5
72	81	75	80	56	61	78	88	90	87
10+Q	10+T	10+U	10+X	11+A	11+F	11+I	11+V	11+W	3 cx
0.3+3	0.3+50	03+50	0.3+50	1+5	1+5	1+10	1+50	1+50	1
84	62	65	64	82	87	92	70	65	42
5 cx	7 cx	8 cx	10 cx	11 cx	A cx	F cx	I cx	M cx	Q cx
0.5	0.3	0.3	0.3	1	5	5	10	0.5	3
39	33	40	48	52	31	42	45	36	41
T cx	U cx	V cx	W cx	X cx					
50	50	50	50	50					
0	0	0	0	0					

[0102] Now, it shall be specifically explained in reference to test examples that the more reliable integrated control of disease injuries by suppressing pathogenic development even after host plants have been inflicted by pathogenic fungi (therapeutic effect) and also by inhibiting the infection by pathogenic fungi over an extended time period (residual effect) can be provide by using the composition of active compounds according to the present invention, rather than by individually using respective active compounds.

[0103] Test Example 4: Integrated effects with the therapeutic effect and residual effect/Testing the effect of protecting cucumbers from downy mildew.

Ten (10) cucumber seeds (the kind: Sagami Hampaku) were sown in every 9cm×9cm sized PVC-made pot for 7-day growth in a greenhouse chamber, and cucumber seedlings having developed cotyledons were used as test plants. The test plants were inoculated with a conidia spore suspension of Pseudoperonospora cubensis by spraying, and then were immediately placed in a moist chamber kept at 20C to promote the infection. Twenty four (24) hours later, the test plants were moved from the moist chamber to a greenhouse chamber. After air drying, compositions according to the present invention, as prepared after the preparatory examples 8 and 29 were diluted with water to predetermined concentrations of active components for respectively spraying 15mL a pot, and a disease was caused to be developed. Fourteen (14) days later, the pathogenic fungus was inoculated again for intrusion according to the same method as in the earlier inoculation, and disease was caused to be developed in a greenhouse chamber. During the test period, sampled foliage leaves were removed at appropriate timing, and a liquid fertilizer was applied to the root section. After the lapse of a predetermined time since chemical applications, the disease development at each cotyledon was examined in the same manner as in Example 1, and based on the observed index values, levels of injuries and control factors were derived in the same manner as in Example 1. Test results are shown in Table 5.

[0104] [Table 5]

test compounds			3+A	3+B	3+D	3+E	3+F	3+G	3+H
active component conc., ppm			10+1000	10+1000	10+1000	10+1000	10+1000	10+1000	10+1000
control factor after 7 days, %			82	78	81	84	80	81	77
control factor after 21 days, %			79	70	75	80	74	76	69
3+L	8+A	8+B	8+D	8+E	8+F	8+G	8+H	8+L	10+A
10+1000	10+1000	10+1000	10+1000	10+1000	10+1000	10+1000	10+1000	10+1000	10+1000
74	92	90	93	91	89	92	93	92	100
70	86	82	88	81	85	87	86	85	100
10+B	10+D	10+E	10+F	10+G	10+H	10+L	3 cx	8 cx	10 cx
10+1000	10+1000	10+1000	10+1000	10+1000	10+1000	10+1000	10	10	10
100	99	98	100	100	98	99	72	85	96
100	92	91	100	100	90	93	12	18	23
A cx	B cx	D cx	E cx	F cx	G cx	H cx	L cx		
1000	1000	1000	1000	1000	1000	1000	1000		
0	0	0	0	0	0	0	0		
0	0	0	0	0	0	0	0		

[0105] Test Example 5: Integrated effects with the therapeutic effect and residual effect/Testing the effect of protecting tomatoes from late blight.

Tomato seedlings (the kind: Ponte Rosa) were transplanted one by one in 12cm diameter unglazed pots for growth in a greenhouse chamber, and tomato seedlings developed to have 6~7 compound leaves were used as test plants. The test plants were inoculated with a swarm ascus suspension of Phytophthora infestans by spraying, and then were immediately placed in a moist chamber kept at 20C to promote the infection. Eight (8) hours later, the test plants were moved from the moist chamber to a greenhouse chamber. After air drying, compositions according to the present invention, as prepared after the preparatory example 9 were diluted with water to predetermined concentrations of active components for respectively spraying 20mL a pot, and a disease was caused to be developed in the greenhouse chamber. Fourteen (14) days later, the pathogenic fungus was inoculated again according to the same method as in the earlier inoculation and was allowed for intrusion and disease development in the moist chamber. After the lapse of a predetermined time since chemical applications, the disease development at each leaflet was examined in the

same manner as in Example 1 to derive control factors. Test results are shown in Table 6.

[0106] [Table 6]

test compounds			10+A	10+B	10+D	10+E	10+F	10+L
active component conc., ppm			10+1500	10+1500	10+1500	10+1500	10+1500	10+1500
control factor after 7 days, %			100	100	100	100	100	100
control factor after 21 days, %			100	100	100	100	100	100
10 cx	A cx	B cx	D cx	E cx	F cx	L cx		
10	1500	1500	1500	1500	1500	1500		
100	0	0	0	0	0	0		
42	0	0	0	0	0	0		

[0107] Test Example 6: Integrated effects with the therapeutic effect and residual effect/Testing the effect of protecting grapes from downy mildew

Grape nursery stocks (the kind: Kyoho) planted in 12cm diameter unglazed pots were pruned and grown in a greenhouse chamber, and the grape nursery stocks developed to have 4~5 leaves were used as test plants. The test plants were inoculated with a conidia spore suspension of Plasmopara viticola by spraying, then, immediately placed in a moist chamber kept at 20°C to promote intrusion. Twenty four (24) hours later, the test plants were moved from the moist chamber to a greenhouse chamber, and after air drying, compositions according to the present invention as prepared after the preparatory example 8 were diluted with water to predetermined concentrations of active components for respectively spraying 20mL a pot and causing the development of a disease in the greenhouse chamber. Fourteen (14) days later, the pathogenic fungus was inoculated again according to the same method as in the earlier inoculation and was allowed for intrusion and subsequent disease development in the greenhouse chamber. After the lapse of a predetermined time since chemical applications, the test plants were again placed in a moist chamber kept at 20°C for 24 hours so that conidia spores could be formed. The size of infected areas including leaf positions where

conidia spores were formed and lesion positions with brown spots and brown pustules were examined according to the standard of infection index described in Example 1, and levels of injuries and control factors were derived in the same manner as in Example 1. Test results are shown in Table 7.

[0108] [Table 7]

test compounds			10+A	10+B	10+D	10+E	10+F	10+L
active component conc., ppm			15+1500	15+1500	15+1500	15+1500	15+1500	15+1500
control factor after 7 days, %			100	100	100	100	100	100
control factor after 21 days, %			100	100	100	100	100	100
10 cx	A cx	B cx	D cx	E cx	F cx	L cx		
15	1500	1500	1500	1500	1500	1500		
100	0	0	0	0	0	0		
36	0	0	0	0	0	0		

[0109] Test Example 4: Testing the effect of protecting cucumbers from downy mildew (phenyl amide-resistant fungus).

Ten (10) cucumber seeds (the kind: Sagami Hampaku) were sown in every 9cm×9cm sized PVC-made pot for 7-day growth in a greenhouse chamber, and cucumber seedlings having developed cotyledons were used as test plants. Compositions according to the present invention, as prepared after the preparatory examples 18 and 33 were diluted with water to predetermined concentrations for respectively spraying at the ratio of 15mL a pot. After air drying, the test plants were inoculated with a conidia spore suspension of Pseudoperonospora cubensis by spraying, and then were immediately placed in a moist chamber kept at 20C, for 24 hours. Later, the test plants were moved into a greenhouse chamber, and the infection level of each cotyledon was examined according to the below described infection index. Based on the observed index values, levels of injuries and control factors were derived. Test results are shown in Table 8.

[0110] [Table 8]

test compounds			3+M	3+N	5+M	5+N	7+M	7+N	10+M
active component conc. ppm			1+0.5	1+0.5	0.5+0.5	0.5+0.5	0.5+0.5	0.5+0.5	0.5+0.5
control factor, %			85	82	79	81	86	90	87
10+N	11+M	11+N	M cx	M cx	N cx	N cx			
0.5+0.5	1+0.5	1+0.5	3	0.5	3	0.5			
85	82	85	0	0	0	0			

[0111]

[Advantages of the Invention] A fungicide composition for agricultural and horticultural uses exhibits excellent activity of controlling various pathogenic fungi at a low dosage, and particularly exhibits excellent activity of controlling Phycomycetes, which causes downy mildew to cucumbers, late blight of tomatoes, downy mildew to grapes, late blight to potatoes, etc. Also, it exhibits excellent post-infection fungicidal activity with a fungicidal compound for agricultural and horticultural uses which has low post-infection fungicidal activity, exhibits outstanding activity of controlling chemical-resistant fungi with a fungicidal compound for agricultural and horticultural uses having a control activity lowered by chemical-resistant fungi, and exhibits extremely high activity of controlling Phycomycetes with a fungicidal compound for agricultural and horticultural uses having the poor or no activity of controlling Phycomycetes. Furthermore, it can effectively inhibit the pathogenic fungus infection over an extended time period and thus can suppress the generation of diseases. It can be also very safely used with useful plants.